

Molecular-Coherent-States and Molecular-Fundamental-States

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to Moshé Flato and André Heslot.

Abstract

New families of Molecular-Coherent-States are constructed by the Perelomov group-method. Each family is generated by a Molecular-Fundamental-State that depends on an arbitrary sequence of complex numbers c_j . Two of these families were already obtained by D.Janssen and by J. A. Morales, E. Deumens and Y. Öhrn. The properties of these families are investigated and we show that most of them are independent on the c_j .

1 Introduction

Since their introduction by Schrödinger [1], the Coherent States of the Harmonic Oscillator (C.S.H.O.) were extensively studied and used in many branches of physics [2]. These states satisfy numerous properties, let us recall some of them :

- 1) The C.S.H.O. constitute an (overcomplete) basis of non orthogonal vectors of the Hilbert Space of the states of the harmonic oscillator \mathcal{H} .
- 2) On this basis, the vectors of \mathcal{H} are realized as entire analytical functions of a complex variable.
- 3) The C.S.H.O. are eigenvectors of the annihilation operator.

- 4) They minimize the uncertainty relations.
- 5) The mean values of the position and of the momentum on the C.S.H.O. evolve in time like the corresponding classical quantities.
- 6) The C.S.H.O. have the temporal stability.
- 7) They are generated by the Heisenberg-Weyl group.

Their generalizations to others systems are constructed in order to verify some of these properties, 1 and 2 being always required.

A fruitful generalization originating from the property 7 was given by Perelomov who defined Coherent States related to other Lie Groups [3]. Applying the group-method to $SU(2)$ [4], he constructed the Spin-Coherent-States studied by Radcliffe [5][6].

In [7] and [8], Coherent-States (C.S.) were found for the quantum mechanical top and for the description of molecular-rotations. These states fulfill the two requirements 1 and 2 and are proved to satisfy the properties 4 and 5. We claim that the proof, based on some relations satisfied by these C.S., is not valid because the property 6 is not fulfilled, i.e. a rotor in a C.S. introduced in [7] and [8] does not remain in a C.S. when time evolves. We come back in detail on this point in the following.

The main interest of the C.S. introduced in [7] and [8] and of the Spin-Coherent-States is to constitute a suitable basis in various applications : asymmetric top [13], forced rotation model [7], time dependent electron nuclear dynamics [8], partition function in a magnetic field [5], spin relaxation process [14]... The C.S. are not unique and the purpose of this paper is to construct new families of C.S. generalizing the states introduced in [7] and [8], to study and compare their properties.

To begin, in Section (2), we recall some well-known properties of the quantum rigid body in order to fix the notations. In section (3), we define Molecular-Coherent-States (denoted in the following M.C.S.) as the result of the action of group-operators on a Molecular-Fundamental State (denoted in the following M.F.S.). The Lie Group acting is $SU(2) \otimes SU(2)$ and the M.F.S. is a generalization of the fundamental vector used in [3]. A M.F.S. is characterized by a sequence of complex numbers c_j , that must verify two conditions in order that the M.C.S. satisfy the requirements 1 and 2. A large arbitrariness remains in the choice of the M.F.S., but once this choice is done, the set of M.C.S. is uniquely defined. The C.S. defined in [7] and [8] are recovered for two specific sequences of c_j .

In section (4), we give the characteristic properties of the M.F.S., the main result is that the choice of the c_j does not play a prominent part in this

study.

In section (5), all the results of the previous section are transformed by the action of the group to set up the list of the characteristic properties of the M.C.S.. The Z -representation is tackled and the representation of the angular momentum as differential operators is given.

The conclusions are contained in the last section and the appendices give some complements : the realization of the angular momentum on the functions of the Euler angles and the representation of the bi-tensors on the canonical basis.

2 Quantum Rigid Body

2.1 Laboratory and Molecular-Components of the Angular Momentum

For any quantum system, the components $J_{(0,1,2)}^L$ of its angular momentum \vec{J} , on a set of three mutually orthogonal laboratory-fixed axes are the generators of the rotation group and verify the $su(2)$ -algebra commutation relations :

$$[J_+^L, J_-^L] = 2J_0^L, \quad [J_0^L, J_\pm^L] = \pm J_\pm^L \quad (1)$$

where the spherical coordinates are defined by $J_\pm \equiv J_1 \pm iJ_2$.

The top-hamiltonian reads

$$H = \sum_{i=0}^2 A_i (J_i^M)^2 \quad (2)$$

where $J_{(0,1,2)}^M$ are the components of the angular momentum \vec{J} on a set of three mutually orthogonal axes moving with the system under a rotation. The rotational constants A_i , inverse of the moments of inertia, characterize the symmetry of the molecule. Writing the molecular-components J^M as the scalar product of \vec{J} with a vector and using the characteristic commutation relations of J^L with the laboratory-components of a vector, we easily prove that the J^M satisfy the following commutation relations

$$[J_+^M, J_-^M] = -2J_0^M, \quad [J_0^M, J_\pm^M] = \mp J_\pm^M \quad (3)$$

and that they commute with all the laboratory-components, symbolically :

$$[J^L, J^M] = 0 \quad (4)$$

The Lie algebra \mathcal{A} , generated by the J^L and the J^M is $su(2) \otimes su(2)$ with the constraint $J^2 = \sum_{i=0}^2 (J_i^L)^2 = \sum_{i=0}^2 (J_i^M)^2$.

Up to now, the rotations considered are the rotations of the body and of the molecular-frame that keep fixed the laboratory-frame; they correspond to unitary transformations R_L of the states and observables of the quantum system

$$R_L(\alpha_L, \beta_L, \gamma_L) = \exp(-i\alpha_L J_0^L) \exp(-i\beta_L J_2^L) \exp(-i\gamma_L J_0^L) \quad (5)$$

Similarly, we can consider rotations of the body and of the laboratory-frame that keep fixed the molecular-frame; the unitary operator R_M associated to these rotations are given by

$$R_M(\alpha_M, \beta_M, \gamma_M) = \exp(-i\alpha_M J_0^M) \exp(i\beta_M J_2^M) \exp(-i\gamma_M J_0^M) \quad (6)$$

Due to (4), the laboratory-rotations, R_L , and the molecular-rotations, R_M , commute.

2.2 Representation of J^L and J^M

The eigenvectors of the three operators J^2, J_0^L, J_0^M constitute the basis of the space of the canonical representation. We have :

$$\begin{aligned} J^2 |j, k, m\rangle &= j(j+1) |j, k, m\rangle, & j &= 0, \frac{1}{2}, 1, \dots \\ J_z^L |j, k, m\rangle &= m |j, k, m\rangle, & m &= -j, -j+1, \dots, j \\ J_z^M |j, k, m\rangle &= k |j, k, m\rangle, & k &= -j, -j+1, \dots, j \end{aligned} \quad (7)$$

When j is fixed, the states $|j, k, m\rangle$ span the $(2j+1)^2$ dimensional Hilbert space h_j .

The action of the operators on the canonical basis is given by :

$$\begin{aligned} J_\pm^L |j, k, m\rangle &= \sqrt{(j \mp m)(j \pm m + 1)} |j, k, m \pm 1\rangle \\ J_\pm^M |j, k, m\rangle &= \sqrt{(j \pm k)(j \mp k + 1)} |j, k \mp 1, m\rangle \end{aligned} \quad (8)$$

Let α, β, γ be the Euler angles relating the laboratory-frame and the molecular-frame and satisfying, by convention,

$$0 \leq \alpha < 2\pi, \quad 0 \leq \beta \leq \pi, \quad -\pi \leq \gamma < \pi. \quad (9)$$

It is well known that the canonical representation can be realized on the space of functions, $\mathcal{C}(\alpha, \beta, \gamma)$, and that j must be an integer number in order that the wave functions of a rigid molecule be single valued (see appendix (7.1)).

In the following, we don't restrict to this case and we construct the M.C.S. spanning either $\mathcal{H}_{\frac{1}{2}} \equiv \oplus_{(j=0, \frac{1}{2}, 1, \dots)} h_j$ or $\mathcal{H}_1 \equiv \oplus_{(j=0, 1, \dots)} h_j$.

2.3 Bi-tensors

The components of a bi-tensor operator commute between themselves and transform under the laboratory or the molecular-rotations according to the formulas [15]

$$R_L T_{q,q'}^{j,j'} R_L^{-1} = \sum_{k'=-j'}^{j'} T_{q,k'}^{j,j'} R_{k'q'}^{j'}, \quad R_M T_{q,q'}^{j,j'} R_M^{-1} = \sum_{k=-j}^j T_{k,q'}^{j,j'} R_{kq}^j \quad (10)$$

where

$$R_{mm'}^j(\alpha, \beta, \gamma) = \exp(-i\alpha m) \exp(-i\gamma m') d_{mm'}^j(\beta) \quad (11)$$

and

$$\begin{aligned} d_{m'm}^j(\beta) \equiv & \sqrt{(j-m')!(j+m')!(j-m)!(j+m)!} \left(\tan\left(\frac{\beta}{2}\right) \right)^{m-m'} \left(\cos\left(\frac{\beta}{2}\right) \right)^{2j} \\ & \times \sum \frac{(-1)^n}{n!(j-m-n)!(j+m'-n)!(m-m'+n)!} \left(\tan\left(\frac{\beta}{2}\right) \right)^{2n} \end{aligned} \quad (12)$$

The resulting commutation relations read:

$$\begin{aligned} [J_{\pm}^L, T_{q,q'}^{j,j'}] &= \sqrt{(j' \mp q')(j' \pm q' + 1)} T_{q,q' \pm 1}^{j,j'}, & [J_0^L, T_{q,q'}^{j,j'}] &= q' T_{q,q'}^{j,j'} \\ [J_{\mp}^M, T_{q,q'}^{j,j'}] &= \sqrt{(j \mp q)(j \pm q + 1)} T_{q \pm 1, q'}^{j,j'}, & [J_0^M, T_{q,q'}^{j,j'}] &= q T_{q,q'}^{j,j'} \end{aligned} \quad (13)$$

The hermitean adjoint is defined by

$$(T^\dagger)_{qq'}^{jj'} = (-1)^{q-q'} T_{-q-q'}^{jj'} \quad (14)$$

We easily verify that $(-\frac{1}{\sqrt{2}}J_+^L, J_0^L, \frac{1}{\sqrt{2}}J_-^L)$ is a bi-tensor $J^{0,1}$ and that $(-\frac{1}{\sqrt{2}}J_-^M, J_0^M, \frac{1}{\sqrt{2}}J_+^M)$ is a bi-tensor $J^{1,0}$.

In the following, we call bi-spinor S the bi-tensor $T^{\frac{1}{2},\frac{1}{2}}$ and bi-vector V the bi-tensor $T^{1,1}$. The components of S and V are represented on the canonical basis in the Appendix 7.2.

3 Coherent States

3.1 Definitions

Definition Let $c_0 \neq 0, c_{\frac{1}{2}}, c_1 \dots$, be an arbitrary sequence of complex numbers, a Molecular-Fundamental-State (M.F.S.), is a state of $\mathcal{H}_{\frac{1}{2}}$ of the form :

$$|z\rangle \equiv \sum_j c_j z^j |j, -j, -j\rangle, \quad z \in C, \quad \sum_j \equiv \sum_{j=0, \frac{1}{2}, 1, \dots} \quad (15)$$

The M.F.S. is the analogous of the fundamental vector generating the Spin C.S. in [3] and constitutes the main ingredient of the group-construction of the M.C.S. proposed in this paper. In order that the M.F.S. belongs to the Hilbert space $\mathcal{H}_{\frac{1}{2}}$, the coefficients c_j and the complex variable z must satisfy the following condition

$$\langle z | z \rangle \equiv N(|z|^2) = \sum_j |c_j|^2 |z|^{2j} < \infty \quad (16)$$

Let us remark that $|z\rangle$ belongs to \mathcal{H}_1 , if $c_j = 0$ when j take half-integer values.

Applying the group-method [3] to the group $SU(2) \otimes SU(2)$, we define

Definition The Molecular-Coherent-States are the states resulting from the action of the laboratory-rotations R_L and of the molecular-rotations R_M upon the M.F.S.(15) and spanning $\mathcal{H}_{\frac{1}{2}}$ (or \mathcal{H}_1 when $|z\rangle \in \mathcal{H}_1$).

Let us denote

$$D_L(\zeta_L) = e^{\zeta_L J_+^L} e^{\eta_L J_0^L} e^{-\bar{\zeta}_L J_-^L}, \quad \eta_L = \ln(1 + |\zeta_L|^2) \quad (17)$$

Writing $R_L(\alpha_L, \beta_L, \gamma_L)$ as the product of $D_L(-\tan \frac{\beta_L}{2} e^{-i\alpha_L})$ by $e^{-i(\alpha_L + \gamma_L)J_0^L}$, we notice that the last term of this product transforms the M.F.S. $|z\rangle$

into $|ze^{i(\alpha_L + \gamma_L)}>$, and therefore that two laboratory-rotations only differing by this last term give the same M.C.S.. The same holds for the molecular-rotations.

Therefore, analogously to $SU(2)$, a M.C.S. system is constructed by applying on the M.F.S.(15) the operators D_L defined in (17) and D_M defined by :

$$D_M(\zeta_M) = e^{\zeta_M J_-^M} e^{\eta_M J_0^M} e^{-\overline{\zeta_M} J_+^M}, \quad \eta_M = \ln(1 + |\zeta_M|^2) \quad (18)$$

A M.C.S. then is of the form

$$|Z> = D_L(\zeta_L) D_M(\zeta_M) |z> \quad (19)$$

Obviously, the norms of $|Z>$ and $|z>$ are both equal to $\sqrt{N(|z|^2)}$ and the M.C.S. exist if the sequence c_j and the complex parameter z verify (16).

The explicit calculation of (19) gives the decomposition of the M.C.S. on the canonical basis

$$|Z> = \sum_{jkm} \sigma_k^j \sigma_m^j \zeta_L^{j+m} \zeta_M^{j+k} z^j c_j (1 + |\zeta_L|^2)^{-j} (1 + |\zeta_M|^2)^{-j} |jkm> \quad (20)$$

with the notations

$$\sum_{jkm} \equiv \sum_{j=0}^{\infty} \sum_{m=-j}^j \sum_{k=-j}^j \quad \text{and} \quad \sigma_k^j \equiv \sqrt{\frac{(2j)!}{(j-k)!(j+k)!}}. \quad (21)$$

Definition A c-set is a set of M.C.S. defined by (15) and (19) and corresponding to a given sequence c_j .

The parameter $Z = (z, \zeta_L, \zeta_M)$ is such as ζ_L and ζ_M belong to the whole complex plane C , z is eventually restricted by (16).

The scalar product of two M.C.S. of a c-set is given by

$$<Z' | Z> = N \left(\frac{(1 + \overline{\zeta'_L} \zeta_L)^2 (1 + \overline{\zeta'_M} \zeta_M)^2 \overline{z'} z}{(1 + |\zeta_L|^2)(1 + |\zeta_M|^2)(1 + |\zeta'_L|^2)(1 + |\zeta'_M|^2)} \right) \quad (22)$$

We illustrate each step of the present study with eight examples.

♣ In the following examples, all the representations occur in the decomposition of the M.C.S. over the canonical basis.

	$c_j, j \text{ integer or half-integer}$	$N(z ^2)$
1	$\frac{1}{\sqrt{(2j)!}}$	$e^{ z ^2}, \quad \forall z $
2	$\sqrt{\frac{2j+1}{2}} \frac{1}{\sqrt{(2j)!}}$	$\frac{1}{2}(1+ z)e^{ z ^2}, \quad \forall z $
3	$(2j+1)\sqrt{j+1}$	$\frac{3 z ^2+2}{2(1- z)^4}, \quad z < 1$
4	$(2j+1)^{\frac{3}{2}}$	$\frac{ z ^2+4 z +1}{(1- z)^4}, \quad z < 1$

Let us stress that :

The M.C.S. 1 were previously studied by D.Janssen [7] who arbitrarily assumed the values of the c_j . In [11], [10] [12] and [14], these specific values of the c_j are obtained for the linear rotor ($k = 0$) by using the Schwinger's method for the construction of the angular momentum algebra .

The M.C.S.3 and the M.C.S.4 only exist if $|z| < 1$.

♣ In the following examples, the coefficients are such that $c_j = 0$ when j is an half-integer number, the M.C.S. only depend on odd-dimensional representations and belong to \mathcal{H}_1 .

	$c_j, j \text{ integer}$	$N(z ^2)$
5	$\frac{1}{\sqrt{j!}}$	$e^{ z ^2}, \quad \forall z $
6	$\frac{2j+1}{\sqrt{j!}}$	$(4 z ^4 + 8 z ^2 + 1)e^{ z ^2}, \quad \forall z $
7	$(2j+1)\sqrt{j+1}$	$\frac{9 z ^4+14 z ^2+1}{(1- z ^2)^4}, \quad z < 1$
8	$(2j+1)^{\frac{3}{2}}$	$\frac{(1+ z ^2)(z ^4+22 z ^2+1)}{(1- z ^2)^4}, \quad z < 1$

The M.C.S.5 were previously introduced by Jorge A. Morales, Erik Deumens and Yngve Öhrn who analyze the results occuring when the half-integer values of j are discarded from the study of Janssen [8].

The M.C.S.7 and the M.C.S.8 only exist if $|z| < 1$.

3.2 Resolution of unity

We impose that the set of M.C.S. span $\mathcal{H}_{\frac{1}{2}}$ (resp. \mathcal{H}_1) and verify a resolution of unity

$$\frac{1}{\pi^3} \int \int \int dz d\bar{z} \frac{d\zeta_L d\bar{\zeta}_L}{(1+|\zeta_L|^2)^2} \frac{d\zeta_M d\bar{\zeta}_M}{(1+|\zeta_M|^2)^2} f(|z|^2) |Z\rangle\langle Z| = 1 \quad (23)$$

where the operator 1 is the unity in $\mathcal{H}_{\frac{1}{2}}$ (resp. in \mathcal{H}_1). The measures in the ζ_L and ζ_M -integrations are the $SU(2)$ -invariant measures and the measure $f(|z|^2)$ must be determined. The calculation of the expression (23) between two states $|jkm\rangle$ and $|j'k'm'\rangle$ shows that the weight-function $f(x)$ is the Mellin-inverse of the function \hat{f} such as :

$$\int dx f(x) x^j = \frac{(2j+1)^2}{|c_j|^2} \equiv \hat{f}(j), \quad j \in 2N \quad (\text{resp. } j \in N) \quad (24)$$

The existence of $f(x)$ and therefore of the resolution of the identity only depends on the choice of the coefficients c_j .

Result : The resolution of the identity (23) exists if the sequence c_j is such that the function \hat{f} defined by (24) is the Mellin transform of a function f .

A large arbitrariness remains in the choice of the c_j . Reciprocally, any function leading to finite momenta gives a sequence c_j using (24) and then a family of M.C.S. provided that the set of z verifying (16) is not reduced to 0.

Let us remark that we cannot restrict the complex variable z to be on a circle because Formula (24) then implies that $|z|^{2j} |c_j|^2 = (2j+1)^2$ and then that the norm of the M.C.S. is infinite.

In the following table, we give the measure $f(x)$ corresponding to the eight examples illustrating the construction.

	$f(z ^2)$
1	$\frac{1}{2}(z - 1)e^{- z }$
2	$e^{- z }$
3	$\theta(1 - z)$
4	$\frac{\theta(1 - z)}{2 z }$
5	$(4 z ^4 - 8 z ^2 + 1)e^{- z ^2}$
6	$e^{- z ^2}$
7	$\theta(1 - z)$
8	$\frac{\theta(1 - z)}{2 z }$

$\theta(x)$ is the Heaviside-function equal to 1 when $x > 0$ and to 0 when $x < 0$. Let us remark that the measures are strictly positive except $f_1(x)$ and $f_5(x)$, previously obtained by [7] and [8].

Formula (23) implies the existence of a reproducing kernel $\langle Z | Z' \rangle$.

To conclude, due to the existence of a resolution of unity, we are able to decompose any state $|\psi\rangle$ on the overcomplete basis of the M.C.S.. This gives the Z -representation of \mathcal{A} .

3.3 Z-Representation

In the Z -Representation, an arbitrary state of $\mathcal{H}_{\frac{1}{2}}$ of the form $|\psi\rangle = \sum_{jkm} c_{jkm} |jkm\rangle$, corresponds to a continuous function $\psi(Z) \equiv \langle \bar{Z} | \psi \rangle$ of the three complex variables ζ, ζ_L and ζ_M where ζ is defined by

$$\zeta = \frac{\zeta_L \zeta_M z}{(1 + |\zeta_L|^2)(1 + |\zeta_M|^2)} \quad (25)$$

Using the expression (19), we obtain

$$\begin{aligned} \psi(Z) &= \langle \bar{Z} | \psi \rangle \\ &= \sum_{jkm} c_{jkm} \sigma_m^j \sigma_k^j \zeta_L^m \zeta_M^k \bar{c}_j \zeta^j \end{aligned} \quad (26)$$

The function associated to the M.F.S. $|z_0\rangle$ is $N\left(\frac{z_0 z}{\zeta_L \zeta_M}\right)$, where N is the norm-function introduced in (16). In this representation, the components of \vec{J} , obtained by calculating $\langle \bar{Z} | J | j, k, m \rangle$, don't depend on the sequence c_j and take the very simple form :

$$\begin{cases} J_-^L &= \frac{1}{\zeta_L}(\zeta \partial_\zeta + \zeta_L \partial_{\zeta_L}) \\ J_+^L &= \zeta_L(\zeta \partial_\zeta - \zeta_L \partial_{\zeta_L}) \\ J_0^L &= \zeta_L \partial_{\zeta_L} \end{cases} \quad (27)$$

and

$$\begin{cases} J_+^M &= \frac{1}{\zeta_M}(\zeta \partial_\zeta + \zeta_M \partial_{\zeta_M}) \\ J_-^M &= \zeta_M(\zeta \partial_\zeta - \zeta_M \partial_{\zeta_M}) \\ J_0^M &= \zeta_M \partial_{\zeta_M} \end{cases} \quad (28)$$

These expressions can be used to calculate the matrix elements $\langle \bar{Z} | T | Z' \rangle$ when T is a polynomial of the components of \vec{J} they are obtained by differentiating the norm-function. Let us remark that due to the specific ranges of the parameters j, k, m given in (21), the space of the functions occurring in (26) is a subspace of $\mathcal{C}(\zeta, \zeta_L, \zeta_M)$ and that the operators J_+ and J_- are not adjoint in the whole space but only in the subspace.

The expression of $\langle \bar{Z} | S | j, k, m \rangle$ involves coefficients such as \sqrt{j}, \dots that correspond to undefined operators $\sqrt{\zeta} \partial_\zeta$ and then the bi-spinor is not a differential operator. In the Z -representation, all the operators B can be determined by the diagonal elements $\langle \bar{Z} | B | \bar{Z} \rangle$. We give the expressions of these quantities when B is a component of \vec{J} , S and V in Section (5).

To end let us give an application of the Z -representation, the study of the asymmetric-top. Replacing (28) in the Hamiltonian (2), we find that the stationary wave functions satisfy a differential equation in the complex variable ζ_M , this equation was previously obtained and studied by Pavlichenkov [13].

4 Properties of the Molecular-Fundamental-States

The M.C.S. being obtained by the action of rotations upon the M.F.S., the study of their properties is simpler if deduced from the properties of the M.F.S.. Let us stress that in the following, the sequence c_j is not specified, moreover we shall see that, except for explicit calculations, the specific choice of this sequence does not play an important part.

4.1 Action of the angular momentum on the M.F.S.

From now on, we discard the label L or M of the components when the formulas hold in both cases. Let us define the operator Λ by its action on the canonical basis

$$\Lambda | j, k, m \rangle = j | j, k, m \rangle \quad (29)$$

Λ commutes with all the generators of \mathcal{A} . We easily prove that

$$J_0 | z \rangle = -\Lambda | z \rangle, \quad J_-^L | z \rangle = 0, \quad J_+^M | z \rangle = 0 \quad (30)$$

and that

$$\begin{aligned} (J^2 + J_0(1 - J_0)) | z \rangle &= (J_-^L J_+^L + 2J_0) | z \rangle \\ &= (J_+^M J_-^M + 2J_0) | z \rangle = 0 \end{aligned} \quad (31)$$

Let us remark that Relations (30) and (31) do not depend on the sequence c_j .

4.2 Mean values of the angular momentum

We calculate

$$\langle z' | J_{\pm} | z \rangle = \langle z' | J_1 | z \rangle = \langle z' | J_2 | z \rangle = 0 \quad (32)$$

and

$$\langle z' | J_0 | z \rangle = \sum (-j) |c_j|^2 (z\bar{z}')^j = -(z\bar{z}') N'(z\bar{z}') \quad (33)$$

The mean values of the square of the components are given by :

$$\langle z' | 2(J_1)^2 | z \rangle = \langle z' | 2(J_2)^2 | z \rangle = - \langle z' | J_0 | z \rangle \quad (34)$$

and

$$\langle z' | (J_0)^2 | z \rangle = (z\bar{z}') (z\bar{z}') N''(z\bar{z}') + N'(z\bar{z}') \quad (35)$$

It results that

$$\langle z' | J^2 | z \rangle = z\bar{z}' \left(z\bar{z}' N''(z\bar{z}') + 2N'(z\bar{z}') \right) \quad (36)$$

Let $\langle T \rangle_z$ be the expectation value of the operator T in the M.F.S. $|z\rangle$,

$$\langle T \rangle_z \equiv \frac{\langle z | T | z \rangle}{\langle z | z \rangle} \quad (37)$$

from (32) and (33), it results that

$$\begin{aligned} \langle J_{\pm} \rangle_z &= 0 \\ \langle J_0 \rangle_z &= - |z|^2 \frac{N'(|z|^2)}{N(|z|^2)} \\ \langle J^2 \rangle_z &= |z|^2 \left(|z|^2 \frac{N''(|z|^2)}{N(|z|^2)} + 2 \frac{N'(|z|^2)}{N(|z|^2)} \right) \end{aligned} \quad (38)$$

Result : The vector of components $\langle J_{0,1,2}^L \rangle_z$ (resp. $\langle J_{0,1,2}^M \rangle_z$) lies on the x_0 -axis of the laboratory-frame (resp. of the molecular-frame), this property is independent of the choice of the sequence c_j .

We calculate the mean values given in (38) for our eight examples. The cases 1 and 5 were previously obtained in [7] and [8].

	$\langle J_0 \rangle_z$	$\langle J^2 \rangle_z$
1	$-\frac{1}{2} z $	$\frac{1}{4} z (3 + z)$
2	$-\frac{1}{2} z \frac{ z +2}{ z +1}$	$\frac{1}{4} z \frac{ z ^2+6 z +6}{ z +1}$
3	$-\frac{1}{2} z \frac{9 z +11}{(1- z)(3 z +2)}$	$\frac{1}{4} z \frac{9 z ^2+58 z +33}{(1- z)^2(3 z +2)}$
4	$- z \frac{ z ^2+7 z +4}{(1- z)(z ^2+4 z +1)}$	$ z \frac{6(z ^2+3 z +1)}{(1- z)^2(z ^2+4 z +1)}$
5	$- z ^2$	$ z ^2 (z ^2 + 2)$
6	$- z ^2 \frac{4 z ^4+16 z ^2+9}{4 z ^4+8 z ^2+1}$	$ z ^2 (z ^2 + 2) \frac{4 z ^4+24 z +9}{4 z ^4+8 z ^2+1}$
7	$- z ^2 \frac{6(3 z ^4+10 z ^2+3)}{(1- z ^2)(9 z ^4+14 z ^2+1)}$	$ z ^2 \frac{6(3 z ^6+32 z ^4+39 z ^2+6)}{(1- z ^2)^2(9 z ^4+14 z ^2+1)}$
8	$- z ^2 \frac{ z ^6+49 z ^4+115 z ^2+27}{(1- z ^4)(z ^4+22 z ^2+1)}$	$ z ^2 \frac{6(9 z ^4+62 z ^2+9)}{(1- z ^2)^2(z ^4+22 z ^2+1)}$

4.3 Uncertainty Relations

The uncertainty relations read

$$\langle (\Delta J_1)^2 \rangle_z \langle (\Delta J_2)^2 \rangle_z \geq \frac{1}{4} \langle J_0 \rangle_z^2, \quad \text{circ.perm.} \quad (39)$$

where the fluctuation of the operator T is defined by $\Delta T = T - \langle T \rangle_z$. From (34), we get :

$$\langle (\Delta J_1)^2 \rangle_z \langle (\Delta J_2)^2 \rangle_z = \frac{1}{4} \langle J_z \rangle_z^2 \quad (40)$$

The M.F.S. minimize one of the uncertainty relations. The two others are minimum if

$$\begin{aligned} 0 &= \langle (\Delta J_1)^2 \rangle_z \langle (\Delta J_0)^2 \rangle_z = \langle (\Delta J_2)^2 \rangle_z \langle (\Delta J_0)^2 \rangle_z = \\ &= \frac{(z\bar{z}')^2 N'(z\bar{z}')}{2(N(z\bar{z}'))^3} \left(N(z\bar{z}') (z\bar{z}') N''(z\bar{z}') + N'(z\bar{z}') \right) - z\bar{z}' (N'(z\bar{z}'))^2 \end{aligned} \quad (41)$$

that gives $xN(x)N''(x) - x(N'(x))^2 + N(x)N'(x) = 0$. The solution of this equation of the form (16) is the monomial x^l , with $2l \in N$. It results that the sequence c_j is restricted to one element $c_j = \delta_{jl}$ and that the M.C.S. span h_l , the resolution of unity does not exist in $\mathcal{H}_{\frac{1}{2}}$ (or \mathcal{H}_1).

Result : The M.F.S. minimize one, and only one, of the uncertainty relations, this property is independent of the choice of the sequence c_j .

4.4 Equation of motion

When the evolution is defined by the hamiltonian (2), the molecular-components of the angular momentum, in the Heisenberg representation, are time-dependent whereas their mean values on the M.F.S. are time-independent. The mean values of the angular momentum do not evolve as the classical angular velocity of the rotor.

4.5 Expectation values of the bi-spinor S and of the bi-vector V

Using the representation (75) and (74), we easily prove that $\langle S_{-+} \rangle_z$ and $\langle S_{+-} \rangle_z$ are equal to 0 for all sequence c_j and that

$$\langle z | S_{--} | z \rangle = \sum_j \bar{c}_{j+\frac{1}{2}} c_j \bar{z}^{j+\frac{1}{2}} z^j \sqrt{\frac{2j+1}{2j+2}} = \overline{\langle z | S_{++} | z \rangle} \quad (42)$$

Result : The $\langle S_{qq'} \rangle_z$ form a 2×2 diagonal-matrix that reduces to the 0-matrix when the representation space is \mathcal{H}_1 , this result is independent on the sequence c_j .

Only the explicit expressions (42) of the diagonal elements depend on the sequence. In the example 4, the diagonal element are explicitly calculated : $\langle S_{--} \rangle_z = 2\bar{z}^{\frac{1}{2}} \frac{1+2|z|}{|z|^2+4|z|+1}$.

The mean values of the operators $V_{qq'}$ on the M.F.S. are calculated by utilizing the expressions (76 \cdots 80), the non-diagonal elements $\langle V_{qq'} \rangle_z$ are equal to 0 and the diagonal elements are given by

$$\begin{aligned}
\langle z | V_{--} | z \rangle &= \sum_j \bar{c}_{j+1} c_j \bar{z}^{j+1} z^j \sqrt{\frac{2j+1}{2j+3}} \\
&= \overline{\langle z | V_{++} | z \rangle}
\end{aligned} \tag{43}$$

$$\langle z | V_{00} | z \rangle = - \sum_j \frac{j}{j+1} |c_j|^2 |z|^{2j}$$

Result : $\langle V \rangle_z$ is a 3×3 diagonal-matrix, two of the diagonal elements are complex conjugate and the third one is real; this result does not depend on the sequence c_j .

Only the explicit expressions of the diagonal elements depend on the choice of the sequence. In the examples 4 and 8, the calculations of (43) give

	$\langle V_{--} \rangle_z$	$\langle V_{00} \rangle_z$
4	$\bar{z} \frac{- z ^2 + 4 z + 3}{ z ^2 + 4 z + 1}$	$\frac{ z ^2(-2 z ^3 + z ^2 - 6 z + 1) - 2(\log(1- z) - z)}{ z ^2(1- z)^4}$
8	$\bar{z} \frac{- z ^8 + 8 z ^6 + 110 z ^4 + 240 z ^2 + 27}{(1- z ^4)(z ^4 + 22 z ^2 + 1)}$	$\frac{-19 z ^8 + 5 z ^6 - 41 z ^4 + 7 z ^2 - (1- z ^2)^4 \log(1- z ^2)}{ z ^2(1+ z ^2)(z ^4 + 22 z ^2 + 1)}$

(44)

Similar results hold for bi-tensors of higher equal rank.

To end, let us stress that it is difficult to study the uncertainty relations between the angular momentum and the bi-tensors S and V , and in particular to find the sequence c_j that minimizes any of them.

In the following section, we show that all the properties of the M.F.S. have a counterpart for the M.C.S.

5 Properties of the Coherent States

5.1 Action of the angular momentum on the M.C.S.

First, we remark that a M.C.S. is not transformed into a M.C.S. by the components of \vec{J} . The operator $D_L(\zeta_L)D_M(\zeta_M)$ transforms the angular momentum \vec{J} in a vector, the laboratory and molecular-components of which

are :

$$D_L(\zeta_L)J_{q'}^L D_L^{-1}(\zeta_L) \equiv J_{q'}^L(\zeta_L), \quad D_M(\zeta, M)J_q^M D_M^{-1}(\zeta_M) \equiv J_q^M(\zeta_M) \quad (45)$$

From (19) and (30), we obtain :

$$J_0^L(\zeta_L) | Z \rangle = J_0^M(\zeta_M) | Z \rangle = -\Lambda | Z \rangle \quad (46)$$

Let us remark that the action of the operators $J_0^L(\zeta_L)$ and of $J_0^M(\zeta_M)$ transforms the set of M.C.S. associated to the sequence c_j into the set of M.C.S. associated to the sequence jc_j and that these two sets of M.C.S. correspond to the same domain of the complex plane z . Formula (46) can be written on the form :

$$\begin{aligned} \left(\frac{1}{2}e^{-i\varphi_L} \sin \theta_L J_+^L + \cos \theta_L J_0^L + \frac{1}{2}e^{i\varphi_L} \sin \theta_L J_-^L \right) | Z \rangle &= -\Lambda | Z \rangle \\ = (\cos \varphi_L \sin \theta_L J_1^L + \sin \varphi_L \sin \theta_L J_2^L + \cos \theta_L J_0^L) | Z \rangle &= (\vec{n}^L(\zeta_L) \cdot \vec{J}) | Z \rangle \end{aligned} \quad (47)$$

An analogous relation holds for the molecular-components of \vec{J} . We write these relations in a more compact form :

$$\left(\Lambda + \vec{n}^M(\zeta_M) \cdot \vec{J} \right) | Z \rangle = \left(\Lambda + \vec{n}^L(\zeta_L) \cdot \vec{J} \right) | Z \rangle = 0 \quad (48)$$

The operator $(\vec{J} \cdot \vec{n}^L(\zeta_L))$ (resp. $\vec{n}^M(\zeta_M) \cdot \vec{J}$) is the projection of the angular momentum \vec{J} on the vector $\vec{n}^L(\zeta_L)$ (resp. $\vec{n}^M(\zeta_M)$), the laboratory-components (resp. molecular-components) of which are $(\cos \varphi_L \sin \theta_L, \sin \varphi_L \sin \theta_L, \cos \theta_L)$ (resp. $(\cos \varphi_M \sin \theta_M, -\sin \varphi_M \sin \theta_M, \cos \theta_M)$). This vector $\vec{n}^L(\zeta_L)$ (resp. $\vec{n}^M(\zeta_M)$) is the transformed of the unit vector of the x_0 -axis of the laboratory (resp. molecular) frame by $D_L(\zeta_L)$ (resp. $D_M(\zeta_M)$).

Result : The projections of the angular momentum \vec{J} on the two vectors $\vec{n}^L(\zeta_L)$ and $\vec{n}^M(\zeta_M)$ transform a M.C.S. belonging to some c-set into the same M.C.S. that do not belong to this c-set.

From (31) we get

$$\begin{aligned} J_-^L(\zeta_L) | Z \rangle &= (\zeta_L^2 J_+^L - 2\zeta_L J_0^L - J_-^L) | Z \rangle = 0 \\ J_+^M(\zeta_M) | Z \rangle &= (\zeta_M^2 J_-^M - 2\zeta_M J_0^M - J_+^M) | Z \rangle = 0 \end{aligned} \quad (49)$$

The first relation was already obtained in [3] for the spin C.S..

Remark : All the relations obtained in this subsection are independent on the sequence c_j .

5.2 Laboratory and Molecular-rotations

Let us put the product of the two laboratory-rotations on the form

$$R_L(\alpha_L, \beta_L, \gamma_L) D(\zeta_L) = D_L(R_L.\zeta_L) e^{i\lambda J_0^L} \quad (50)$$

where

$$R_L.\zeta_L = \frac{u_L \zeta_L + v_L}{\overline{u_L} - \overline{v_L} \zeta_L} \text{ and } e^{i\lambda} = \left(\frac{u_L - v_L \overline{\zeta_L}}{\overline{u_L} - \overline{v_L} \zeta_L} \right) \quad (51)$$

We have denoted $u_L = e^{-i\frac{\alpha_L + \gamma_L}{2}} \cos \frac{\beta_L}{2}$ and $v_L = e^{i\frac{\alpha_L - \gamma_L}{2}} \sin \frac{\beta_L}{2}$.

The action of the laboratory-rotation on the M.C.S.(19) result from (50)

$$\begin{aligned} R_L(\alpha_L, \beta_L, \gamma_L) | Z > &= \sum_j c_j z^j \left(\frac{\overline{u_L} - \overline{v_L} \zeta_L}{u_L - v_L \zeta_L} \right)^j D_L^j(R_L.\zeta_L) D_M^j(\zeta_M) | j, -j, -j > \\ &= D_L(R_L.\zeta_L) D_M^j(\zeta_M) | R_L.z > \end{aligned} \quad (52)$$

where

$$R_L.z = z \frac{\overline{u_L} - \overline{v_L} \zeta_L}{u_L - v_L \zeta_L} \quad (53)$$

This result reads

$$R_L(\alpha_L, \beta_L, \gamma_L) | z, \zeta_L, \zeta_M > = | R_L.z, R_L.\zeta_L, \zeta_M > \quad (54)$$

Let us remark that

- the M.F.S. $| z >$ and $| R_L.z >$ correspond to the sequence c_j , the laboratory-rotations $R_L(\alpha_L, \beta_L, \gamma_L)$ transform a M.C.S. of a c-set into a M.C.S. of the same c-set.

- due to the equality $| z | = | R_L.z |$, the norms of the two M.C.S. $| Z >$ and $R_L | Z >$ are equal.

Obviously, we get the analogous result for the action of molecular-rotations R_M that act in one c-set according to the formula :

$$R_M(\alpha_M, \beta_M, \gamma_M) | z, \zeta_L, \zeta_M > = | R_M.z, \zeta_L, R_M.\zeta_M > \quad (55)$$

where the transformed complex variables $R_M.z$ and $R_M.\zeta_M$ are given by the formulas obtained by replacing the label L by M in (51) and (53). The molecular-rotations play a crucial part in the study of the symmetry of the molecule that will be the subject of a forthcoming paper.

5.3 Expectation values of the angular momentum

The rotations transform a bi-tensor $T_{qq'}^{jj'}$ into the operators

$$D_L(\zeta_L)D_M(\zeta_M)T_{qq'}^{jj'}D_L^{-1}(\zeta_L)D_M^{-1}(\zeta_M) \equiv T_{qq'}^{jj'}(\zeta_M, \zeta_L) \quad (56)$$

that satisfy the commutation relations (13) in which the angular momentum is replaced by the transformed angular momentum (45). From (10) and (19), one deduces

$$\begin{aligned} \langle z | T_{qq'}^{jj'} | z \rangle &= \langle Z | T_{qq'}^{jj'}(\zeta_M, \zeta_L) | Z \rangle \\ &= \sum_{k'=-j'}^{j'} \sum_{k=-j}^j \tilde{D}_{qk}^j(\zeta_M) \langle Z | T_{kk'}^{jj'} | Z \rangle D_{k'q'}^{j'}(\zeta_L) \end{aligned} \quad (57)$$

Let $\langle T \rangle_Z$ denote the expectation value of T in the state $| Z \rangle$, namely $\frac{\langle Z | T | Z \rangle}{\langle Z | Z \rangle}$. We have

$$\langle T \rangle_z = \langle T(\zeta_M, \zeta_L) \rangle_Z = \tilde{D}(\zeta_M) \langle T \rangle_Z D(\zeta_L) \quad (58)$$

Let us apply the previous result to the case of the bi-tensor T^{01} and T^{10} . From (58) and (38), we deduce that :

♣ The vector $\langle J_{0,1,2}^L(\zeta_L) \rangle$ lies along the x_0 -axis of the laboratory-frame. Analogously, the vector $\langle J_{0,1,2}^M(\zeta_M) \rangle$ lies along the x_0 -axis of the molecular-frame.

♣ Remembering that $(-\frac{1}{\sqrt{2}}J_+^L, J_0^L, \frac{1}{\sqrt{2}}J_-^L)$ is a bi-tensor $J_{0q'}^{01}$, the calculation of $\langle Z | J_{q'}^L | Z \rangle$ is done by using (58), we obtain

$$\begin{aligned} \langle Z | J_{q'}^{01} | Z \rangle &= \sum_{-1}^1 \langle z | J_{k'}^{01} | z \rangle D_{k'q'}^1(-\zeta_L) \\ \langle J_{q'}^{01} \rangle_Z &= \langle J_0 \rangle_Z D_{0q'}^1(-\zeta_L) \end{aligned} \quad (59)$$

We verify that the vector $\langle J_{q'}^L \rangle_Z$ is parallel to the previously introduced vector $n_{q'}^L(\zeta_L) = D_{0q'}^1(-\zeta_L)$. This result was obtained in [7] and [8], but it is interesting to point out that this result holds for any c-set of M.C.S. Only the length of the vector depends on the choice of coefficients c_j and of the value of z .

♣ Similarly, the calculation of $\langle Z | J_{q'}^M | Z \rangle$ is performed by applying (58) to the bi-tensor $J_{q0}^{10} = (-\frac{1}{\sqrt{2}}J_-^M, J_0^M, \frac{1}{\sqrt{2}}J_+^M)$

$$\begin{aligned}
\langle Z | J_q^{10} | Z \rangle &= \sum_{-1}^1 \langle z | J_k^{10} | z \rangle D_{kq}^1(-\zeta_M) \\
\langle J_q^{10} \rangle_Z &= \langle J_0 \rangle_z D_{0q}^1(-\zeta_M)
\end{aligned} \tag{60}$$

The vectors $\langle J^M \rangle_Z$ and $\vec{n}^M(\zeta_M)$ are parallel. This result does not depend on the c-set considered.

Let us remark that the norms of the vectors $\langle \vec{J}^M \rangle_Z$ and $\langle \vec{J}^L \rangle_Z$ are both equal to the absolute value of $\langle J_0 \rangle_z = -|z|^2 \frac{N'(|z|^2)}{N(|z|^2)}$. The expectation values of the Casimir operator J^2 on the M.F.S. and on the M.C.S. are equal, from (??), we have :

$$\langle J^2 \rangle_Z = |z|^2 \left(|z|^2 \frac{N''(|z|^2)}{N(|z|^2)} + 2 \frac{N'(|z|^2)}{N(|z|^2)} \right) \tag{61}$$

Result : Interpretation of Z

The angles θ_L and φ_L (resp. θ_M and φ_M) define the direction of the vector $\langle J_{q'}^L \rangle_Z$ (resp. $\langle J_q^M \rangle_Z$) in the laboratory (resp. molecular) frame. The modulus $|z|$ and the choice of the c_j are related to the length of these vectors and to the mean values of J^2 .

5.4 Uncertainty Relations

The transformed operators $J_i^L(\zeta_L)$ and $J_i^M(\zeta_M)$ satisfy the same commutation relations (1) and (3) as J_i^L and J_i^M . Therefore their fluctuations obey the same inequalities (39). We easily verify that $\langle (\Delta \vec{J})^2 \rangle_z = \langle (\Delta \vec{J}(\zeta))^2 \rangle_Z$. From the equality (40), we establish that the M.C.S. minimize two uncertainty relations, namely

$$\langle (\Delta J_1^L(\zeta_L))^2 \rangle_Z \langle (\Delta J_2^L(\zeta_L))^2 \rangle_Z = \frac{1}{4} \left(\langle J_0^L(\zeta_L) \rangle_Z \right)^2 \tag{62}$$

and

$$\langle (\Delta J_1^M(\zeta_M))^2 \rangle_Z \langle (\Delta J_2^M(\zeta_M))^2 \rangle_Z = \frac{1}{4} \left(\langle J_0^M(\zeta_M) \rangle_Z \right)^2 \tag{63}$$

The M.C.S. do not minimize the uncertainty relations involving the J_i^L and J_i^M , as studied by [7] [8], but verify two equalities (62) and (63) involving the transformed operators $J_i^L(\zeta_L)$ and $J_i^M(\zeta_M)$. A similar result occurs for the spin C.S.[3].

5.5 Expectation values of the bi-spinor and the bi-vector

It results from Formula (58) that the two matrices $\langle S(\zeta_M, \zeta_L) \rangle_Z$ and $\langle V(\zeta_M, \zeta_L) \rangle_Z$ are diagonal and that

$$\langle Z | S_{qq'} | Z \rangle = \sum_{k' = (-\frac{1}{2}, \frac{1}{2})} \sum_{k = (-\frac{1}{2}, \frac{1}{2})} \tilde{R}_{qk}^{\frac{1}{2}}(-\zeta_M) \langle z | S_{kk'} | z \rangle R_{k'q'}^{\frac{1}{2}}(-\zeta_L) \quad (64)$$

The matrix $\langle S \rangle_Z$ then is the product of one matrix $\tilde{R}^{\frac{1}{2}}(-\zeta_M)$ only depending on ζ_M , one diagonal matrix $\langle S \rangle_z$ only depending on the coefficients c_j and on z , and one matrix $R^{\frac{1}{2}}(-\zeta_L)$ only depending on ζ_L .

Similarly, applying the formula(58) to the bi-vector, we get

$$\langle Z | V_{qq'} | Z \rangle = \sum_{k' = (-1, 1)} \sum_{k = (-1, 1)} \tilde{R}_{qk}^1(-\zeta_M) \langle z | V_{kk'} | z \rangle R_{k'q'}^1(-\zeta_L) \quad (65)$$

Therefore the matrix $\langle V \rangle_Z$ is the product of $\tilde{R}^1(-\zeta_M)$ by the diagonal matrix $\langle V \rangle_z$ and by $R^1(-\zeta_L)$, $\langle V \rangle_z$ depends on the coefficients c_j and on z .

These results can be extended to bi-tensors of higher equal rank.

In conclusion, we have obtained the decomposition of the bi-spinor matrix $\langle S \rangle_Z$ and of the bi-vector matrix $\langle V \rangle_Z$ in terms of three matrices, each of these matrices only depends on one complex variable z , ζ_L or ζ_M .

5.6 Evolution equation

♣ **Rotor** Let us consider a quantum rigid molecule described by the hamiltonian (2). In the Schrödinger representation, the evolution of the M.C.S. $| Z \rangle$ is given by $e^{iHt} | Z \rangle$ that obviously is not a M.C.S. A top in a M.C.S. does not remain in a M.C.S. All the demonstrations of [7] and [8], based on the properties of the M.C.S. are not valid at a time $t \neq 0$. In particular, the expectation values $\langle J_i J_k + J_k J_i \rangle$ are not equal to $2 \langle J_i \rangle \langle J_k \rangle$ when $t \neq 0$ and the evolution equations of the expectation values of the angular momentum are not classical.

Remarks : For the spherical rotor ($A_1 = A_2 = A_3$),

- the M.C.S. corresponding to a sequence c_j become the M.C.S. corresponding to a sequence $c_j e^{it \frac{j(j+1)}{A}}$ during the motion,
- the expectation values of all the components of the angular momentum are constant and then correspond to the classical rotation vector.
- Following [9], we obtain M.C.S. that have the temporal stability by replacing z^j by $|z|^j e^{j\tau(j+1)}$ in (15).

♣ **Temporal stability of (19)**

We look for an Hamiltonian $H_{\mathfrak{h}}$ such that, in the Schrödinger representation, the system is described by the state $|Z(t)\rangle \equiv |z(t), \zeta_L(t), \zeta_M(t)\rangle$. Using the definition (19) of the M.C.S. and the expressions of the components of \vec{J} given in Section (3.3), we prove that the evolution equation of the state is of the form :

$$\begin{aligned} i\partial_t |Z(t)\rangle &= H_{\mathfrak{h}} |Z(t)\rangle \\ &= \left(i(a^L J_+^L - \bar{a}^L J_-^L) + a_0^L J_0^L + i(a^M J_+^M - \bar{a}^M J_-^M) + a_0^M J_0^M \right) |Z(t)\rangle \end{aligned} \quad (66)$$

The complex variables $\zeta_L(t)$ and $\zeta_M(t)$ defining the M.C.S. are related to the coefficients a occurring in the hamiltonian $H_{\mathfrak{h}}$:

$$\dot{\zeta}_L(t) = a^L + \bar{a}^L \zeta_L^2(t) - i a_0^L \zeta_L(t), \quad (67)$$

and

$$\dot{\zeta}_M(t) = a^M + \bar{a}^M \zeta_M^2(t) - i a_0^M \zeta_M(t) \quad (68)$$

The complex variable $z(t)$ must be of the form $z e^{-i\sigma(t)}$ in order that the $H_{\mathfrak{h}}$ be hermitean and $\sigma(t)$ must verify

$$\dot{\sigma}(t) = i \left(a^L \bar{\zeta}_L(t) - \bar{a}^L \zeta_L(t) \right) - a_0^L + i \left(a^M \bar{\zeta}_M(t) - \bar{a}^M \zeta_M(t) \right) - a_0^M \quad (69)$$

Writing $H_{\mathfrak{h}}$ on the form $\sum_i h_i^L J_i^L$, we deduce that this equation describes the motion of a rigid body in a magnetic field h_i^L , that depends on the time through the coefficients a and of the position of the rigid body in the laboratory through the molecular-components J^M .

Let us stress that the state $|Z(t)\rangle$ being a M.C.S., the expectation values of the components of \vec{J} take the form (59) and (60):

$$\langle Z(t) | J_i^L | Z(t) \rangle = \langle J_0 \rangle_z \nu_i^L(t), \quad \langle Z(t) | J_i^M | Z(t) \rangle = \langle J_0 \rangle_z \nu_i^M(t) \quad (70)$$

where the vectors ${}_n\vec{u}^L(t)$ and $\vec{\nu}^M(t)$ verify classical equations of motion. This generalizes the result of Perelomov [4] for $su(2)$.

In the previous reasoning, the c_j are time-independent. When the c_j depend on t , the hamiltonian $H_{\mathfrak{h}}$ contains an extra term that is a function of the Casimir operator J^2 and of t .

6 Conclusion

Molecular-Coherent-States are constructed by transforming Molecular-Fundamental-States by laboratory and molecular-rotations. A M.F.S. is assumed to be a linear combination of the form (15) in which the coefficients c_j have to verify two conditions in order that the M.C.S. satisfy the properties :

- 1) The M.C.S. constitute an (overcomplete) basis of non orthogonal vectors of $\mathcal{H}_{\frac{1}{2}}$ (or eventually \mathcal{H}_1).
- 2) The vectors of $\mathcal{H}_{\frac{1}{2}}$ are realized as continuous functions of three complex variables in section (3.3).

We have established the list of properties of the M.C.S., in analogy to that given in the introduction for the C.S.H.O :

- 3) The four operators defined in (48) and (49) transform the M.C.S. into 0.
- 4) The M.C.S. minimize two uncertainty relations (62) and (63).
- 5) The expectation values of the components of the angular momentum evolve classically for a molecule in a magnetic field.
- 6) For such a quantum system, the temporal stability is verified. When time evolves, a M.C.S. remains a M.C.S.. However, this is not true for the top-hamiltonian (2) contrary to what was claimed by [7] and [8].
- 7) By construction, the M.C.S. are generated by the group of laboratory and molecular-rotations.

We have seen that the prominent part in the group-construction of the M.C.S. is played by the M.F.S.. All the calculations involving M.C.S. are reduced to simpler ones involving M.F.S.. In particular, we easily establish that the matrices of the expectation values of the bi-spinor and the bi-vector are decomposed into the product of two rotations and a diagonal matrix.

To conclude, let us stress the following results :

- the choice of the M.F.S. is the only arbitrariness of the group-construction of the M.C.S.,
 - the fact that the M.F.S. are expressed in terms of the vectors $|j, -j, -j\rangle$ play a crucial part in the establishment of all the properties,
 - these properties are true for any sequence c_j , and we were not able to distinguish and choose a specific sequence and then a more prominent M.F.S..
- Therefore, we can choose in each problem the more convenient basis.

7 Appendix

7.1 Realization in $\mathcal{C}(\alpha, \beta, \gamma)$

In the space of the functions of the Euler angles, $\mathcal{C}(\alpha, \beta, \gamma)$, the laboratory and the molecular-components of the angular momentum take the form :

$$\begin{aligned} J_+^L &= i \exp(i\alpha) \left(\cot \beta \partial_\alpha - i \partial_\beta - \frac{1}{\sin \beta} \partial_\gamma \right) \\ J_-^L &= i \exp(-i\alpha) \left(\cot \beta \partial_\alpha + i \partial_\beta - \frac{1}{\sin \beta} \partial_\gamma \right) \\ J_0^L &= -i \partial_\alpha \end{aligned} \quad (71)$$

and

$$\begin{aligned} J_+^M &= -i \exp(-i\gamma) \left(\cot \beta \partial_\gamma + i \partial_\beta - \frac{1}{\sin \beta} \partial_\alpha \right) \\ J_-^M &= -i \exp(i\gamma) \left(\cot \beta \partial_\gamma - i \partial_\beta - \frac{1}{\sin \beta} \partial_\alpha \right) \\ J_0^M &= -i \partial_\gamma \end{aligned} \quad (72)$$

The states $|j, k, m\rangle$ are represented by the functions

$$\langle \alpha, \beta, \gamma | jkm \rangle = R_{mk}^{*j}(\alpha, \beta, \gamma) = \sqrt{2j+1} \exp(im\alpha) \exp(ik\gamma) d_{mk}^j(\beta) \quad (73)$$

where the R_{mk}^j were defined in (11) and (12). The functions are singled valued if j, k and m are integer numbers.

A M.F.S. is realized as a function of the variable $ze^{-i(\alpha+\gamma)} \cos^2 \frac{\beta}{2}$.

7.2 Representation of S and V

Using the formulas (13) and (8), we obtain the actions of all the components of S on the canonical basis from one of them. We get :

$$\begin{aligned}
S_{+\pm} |j, k, m\rangle &= \frac{\sqrt{(j\pm m+1)(j+k+1)}}{\sqrt{2(j+1)(2j+1)}} |j + \frac{1}{2}, k + \frac{1}{2}, m \pm \frac{1}{2}\rangle \\
&\pm \frac{\sqrt{(j\mp m)(j-k)}}{\sqrt{2j(2j+1)}} |j - \frac{1}{2}, k + \frac{1}{2}, m \pm \frac{1}{2}\rangle,
\end{aligned} \tag{74}$$

$$\begin{aligned}
S_{-\pm} |j, k, m\rangle &= \frac{\sqrt{(j\pm m+1)(j-k+1)}}{\sqrt{2(j+1)(2j+1)}} |j + \frac{1}{2}, k - \frac{1}{2}, m \pm \frac{1}{2}\rangle \\
&\mp \frac{\sqrt{(j\mp m)(j+k)}}{\sqrt{2j(2j+1)}} |j - \frac{1}{2}, k - \frac{1}{2}, m \pm \frac{1}{2}\rangle,
\end{aligned} \tag{75}$$

The components of V act on the canonical basis according to the formulas :

$$\begin{aligned}
V_{-\pm} |j, k, m\rangle &= \frac{\sqrt{(j\pm m+1)(j\pm m+2)(j-k+1)(j-k+2)}}{2(j+1)\sqrt{(2j+1)(2j+3)}} |j + 1, k - 1, m \pm 1\rangle \\
&\mp \frac{\sqrt{(j\mp m)(j\pm m+1)(j-k+1)(j+k)}}{2(j+1)j} |j, k - 1, m \pm 1\rangle \\
&+ \frac{\sqrt{(j\mp m-1)(j\mp m)(j+k-1)(j+k)}}{2j\sqrt{(2j+1)(2j-1)}} |j - 1, k - 1, m \pm 1\rangle
\end{aligned} \tag{76}$$

$$\begin{aligned}
V_{+\pm} |j, k, m\rangle &= \frac{\sqrt{(j\pm m+1)(j\pm m+2)(j+k+1)(j+k+2)}}{2(j+1)\sqrt{(2j+1)(2j+3)}} |j + 1, k + 1, m \pm 1\rangle \\
&\pm \frac{\sqrt{(j\pm m+1)(j\mp m)(j+k+1)(j-k)}}{2(j+1)j} |j, k + 1, m \pm 1\rangle \\
&+ \frac{\sqrt{(j\mp m-1)(j\mp m)(j-k-1)(j-k)}}{2j\sqrt{(2j+1)(2j-1)}} |j - 1, k + 1, m \pm 1\rangle
\end{aligned} \tag{77}$$

$$\begin{aligned}
V_{\pm 0} |j, k, m\rangle &= \frac{\sqrt{(j-m+1)(j+m+1)(j\pm k+1)(j\pm k+2)}}{\sqrt{2}(j+1)\sqrt{(2j+1)(2j+3)}} |j + 1, k \pm 1, m\rangle \\
&\mp \frac{m\sqrt{(j\mp k)(j\pm k+1)}}{\sqrt{2}(j+1)j} |j, k \pm 1, m\rangle \\
&- \frac{\sqrt{(j-m)(j+m)(j\mp k-1)(j\mp k)}}{\sqrt{2}j\sqrt{(2j+1)(2j-1)}} |j - 1, k \pm 1, m\rangle
\end{aligned} \tag{78}$$

$$\begin{aligned}
V_{0\pm} |j, k, m\rangle &= \frac{\sqrt{(j\pm m+1)(j\pm m+2)(j-k+1)(j+k+1)}}{\sqrt{2}(j+1)\sqrt{(2j+1)(2j+3)}} |j+1, k, m\pm 1\rangle \\
&\mp \frac{k\sqrt{(j\mp m)(j\pm m+1)}}{\sqrt{2}(j+1)j} |j, k, m\pm 1\rangle \\
&- \frac{\sqrt{(j\mp m-1)(j\mp m)(j-k)(j+k)}}{\sqrt{2}j\sqrt{(2j+1)(2j-1)}} |j-1, k, m\pm 1\rangle
\end{aligned} \tag{79}$$

$$\begin{aligned}
V_{00} |j, k, m\rangle &= \frac{\sqrt{(j-m+1)(j+m+1)(j-k+1)(j+k+1)}}{(j+1)\sqrt{(2j+1)(2j+3)}} |j+1, k, m\rangle \\
&+ \frac{mk}{(j+1)j} |j, k, m\rangle \\
&+ \frac{\sqrt{(j-m)(j+m)(j-k)(j+k)}}{j\sqrt{(2j+1)(2j-1)}} |j-1, k, m\rangle
\end{aligned} \tag{80}$$

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